

# Full Band Structure Calculation of Two-photon Indirect Absorption in Bulk Silicon

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Degenerate two-photon indirect absorption in silicon is an important limiting effect on the use of silicon structures for all-optical information processing at telecommunication wavelengths. We perform a full band structure calculation to investigate two-photon indirect absorption in bulk silicon, using a pseudopotential description of the energy bands and an adiabatic bond charge model to describe phonon dispersion and polarization. Our results agree well with some recent experimental results. The transverse acoustic/optical phonon-assisted processes dominate.

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Silicon photonics<sup>1-3</sup> has attracted widespread interest due to its potential application in ultracompact all-optical and optical-electronic devices, particularly for use at telecommunication wavelengths near 1.55  $\mu\text{m}$ . Many of these applications are based on the change in index of refraction with intensity, a third-order optical nonlinearity.<sup>4-6</sup> Two-photon absorption, which can plague optical devices because of both immediate nonlinear loss and subsequent loss due to free-carrier absorption, is also a third-order nonlinearity. However, in the interesting wavelength range of 1.2  $\mu\text{m}$  to 1.7  $\mu\text{m}$  the sum of energies from two photons is only sufficient to excite an electron across the indirect band gap, with the assistance of an absorbed or emitted phonon. So indirect degenerate two-photon absorption is of special interest for the development of optical devices,<sup>7-11</sup> and has been the subject of many investigations.<sup>12-20</sup>

Experimentally, the two-photon absorption coefficient  $\beta$  was first measured at 1.06  $\mu\text{m}$  and 100 K by Reintjes and McGroddy,<sup>12</sup> and found to be 1.5 cm/GW. Thereafter a series of experiments at telecommunication wavelengths gave  $\beta$  in the range of 0.44 – 0.9 cm/GW.<sup>6</sup> Bristow *et al.*<sup>15</sup> measured  $\beta$  as a function of photon wavelength and found a strong dependence (see Fig. 1). Anisotropy in the two-photon absorption was also investigated.<sup>16</sup> Theoretically, most studies<sup>18,19</sup> are based on the parabolic band approximation and phenomenological electron-phonon interactions. The exciton effect has also been investigated,<sup>20</sup> but only in quantum wells. Yet a full band structure calculation of the two-photon indirect absorption coefficient is still absent.

In this paper we study the two-photon indirect absorption spectrum in silicon. We perform a full band structure calculation using an empirical pseudopotential model (EPM)<sup>21</sup> for the electronic states, and an adiabatic bond charge model (ABCM)<sup>22</sup> for the phonons, and present the anisotropy and temperature dependence of the absorption. The calculated two-photon absorption coefficient is in good agreement with some recent exper-

imental results.

For an incident laser beam with electric field  $\mathbf{E}(t) = \mathbf{E}_\omega e^{-i\omega t} + c.c.$ , the rate of the carrier density injection is written as

$$\dot{n} = \xi^{abcd} E_\omega^a E_\omega^b (E_\omega^c E_\omega^d)^* . \quad (1)$$

The superscripts indicate Cartesian coordinates, and repeated superscripts are to be summed over;  $\xi^{abcd} = \sum_{\lambda\pm} \xi_{\lambda\pm}^{abcd}$  are the injection coefficients, with  $\xi_{\lambda\pm}^{abcd}$  standing for the contribution from the  $\lambda^{th}$ -branch phonon emission (+) or absorption (−) process. From Fermi's Golden Rule we have

$$\xi_{\lambda\pm}^{abcd} = \frac{2\pi}{\hbar} \sum_{cv\mathbf{k}_c \in i, \mathbf{k}_v} \delta(\varepsilon_{c\mathbf{k}_c} - \varepsilon_{v\mathbf{k}_v} \pm \hbar\Omega_{\mathbf{k}_c-\mathbf{k}_v}^\lambda - 2\hbar\omega) \times \left[ N_{(\mathbf{k}_c-\mathbf{k}_v)\lambda} + \frac{1}{2} \pm \frac{1}{2} \right] W_{c\mathbf{k}_c v\mathbf{k}_v \lambda}^{ab} (W_{c\mathbf{k}_c v\mathbf{k}_v \lambda}^{cd})^* , \quad (2)$$

with the transition matrix elements

$$W_{c\mathbf{k}_c v\mathbf{k}_v \lambda}^{ab} = \left\{ \frac{i}{2} \left( \frac{e}{\hbar\omega} \right)^2 \sum_{nm} \left[ \frac{M_{c\mathbf{k}_c n\mathbf{k}_v; \lambda} v_{nm\mathbf{k}_v}^a v_{mv\mathbf{k}_v}^b}{(\omega_{nv\mathbf{k}_v} - 2\omega)(\omega_{mv\mathbf{k}_v} - \omega)} - \frac{v_{cn\mathbf{k}_c}^a M_{n\mathbf{k}_c m\mathbf{k}_v; \lambda} v_{mv\mathbf{k}_v}^b}{(\omega_{cn\mathbf{k}_c} - \omega)(\omega_{mv\mathbf{k}_v} - \omega)} + \frac{v_{cn\mathbf{k}_c}^a v_{nm\mathbf{k}_c}^b M_{m\mathbf{k}_c v\mathbf{k}_v; \lambda}}{(\omega_{cn\mathbf{k}_c} - \omega)(\omega_{cm\mathbf{k}_c} - 2\omega)} \right] \right\} + \{a \leftrightarrow b\} . \quad (3)$$

Here  $\mathbf{k}_c$  and  $\mathbf{k}_v$  are the electron and hole wavevectors respectively;  $\{a \leftrightarrow b\}$  stands for the same term as in the previous  $\{\}$ , but with the interchange  $a \leftrightarrow b$ . The subscript  $c$  ( $v$ ) is a band index for a conduction (valence) state;  $n, m$  identify intermediate states;  $\varepsilon_{n\mathbf{k}}$  is the electron energy at band  $n$  and wavevector  $\mathbf{k}$ , and  $\hbar\omega_{nm\mathbf{k}} = \varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}}$ . The phonon energy is given by  $\hbar\Omega_{\mathbf{q}\lambda}$  for wavevector  $\mathbf{q}$  and branch  $\lambda$  (transverse acoustic (TA) and optical (TO), and longitudinal acoustic (LA) and optical (LO) branches), and  $N_{\mathbf{q}\lambda}$  is the equilibrium phonon number. The velocity matrix elements are  $v_{nm\mathbf{k}} =$

$\langle n\mathbf{k} | \frac{\partial H_e}{\partial \mathbf{p}} | m\mathbf{k} \rangle$ , with  $H_e$  standing for the single electron Hamiltonian, and the  $M_{n\mathbf{k}_c m\mathbf{k}_v \lambda} = \langle n\mathbf{k}_c | H_\lambda^{ep}(\mathbf{k}_c - \mathbf{k}_v) | m\mathbf{k}_v \rangle$  are matrix elements of the electron-phonon interaction;  $H^{ep} = \sum_{\lambda \mathbf{q}} H_\lambda^{ep}(\mathbf{q})(a_{\mathbf{q}\lambda} + a_{-\mathbf{q}\lambda}^\dagger)$  is the electron-phonon interaction, with  $a_{\mathbf{q}\lambda}$  standing for the phonon annihilation operator.

Due to the  $O_h$  symmetry in bulk silicon, the fourth-order tensor  $\xi^{abcd}$  (with  $\xi^{abcd} = \xi^{bacd}$  and  $\xi^{abcd} = \xi^{abdc}$ ) only has three independent nonzero components:

$$\begin{aligned} \xi^{xxxx} &= \xi^{yyyy} = \xi^{zzzz}, \\ \xi^{xxyy} &= \xi^{xxzz} = \xi^{yyzz} = \xi^{yyxx} = \xi^{zzxx} = \xi^{zzyy}, \\ \xi^{xyxy} &= \xi^{xzzz} = \xi^{yzyz}; \end{aligned} \quad (4)$$

the  $\xi_{\lambda\pm}^{abcd}$  share the same symmetry properties.

For a quantitative calculation, we employ the EPM for the electronic states and the ABCM for the phonon states; all parameters are those used in the calculation of one-photon indirect absorption,<sup>23</sup> where good agreement with experiment has been achieved. Within the pseudopotential scheme we determine the electron-phonon interaction, and then evaluate the matrix elements  $H_\lambda^{ep}(\mathbf{q})$  using the calculated electron and phonon wavefunctions. At zero temperature, the calculated indirect band gap is  $E_{ig} = 1.17$  eV, but it decreases with increasing temperature<sup>24</sup> due to the electron-phonon interaction.<sup>25</sup> Thus we present our results as a function of the excess photon energy  $2\hbar\omega - E_{ig}$ . In the numerical calculation, we use an improved adaptive linear analytic tetrahedral integration method to perform the six-fold integration,<sup>23</sup> and choose the lowest 30 bands as intermediate states to ensure convergence. According to investigations on one-<sup>26</sup> and two-photon<sup>20</sup> indirect gap absorption within the parabolic band approximation, the neglect of the excitonic effects does not change the lineshape at energies more than a few binding energies above the band gap, and we neglect them here. However, since first principle studies of one-photon direct-gap absorption<sup>27</sup> show that the electron-hole interaction plays an important role even for higher photon energies, further work is in order to investigate its role in indirect gap absorption.

We first compare our calculated indirect two-photon absorption coefficient with experiments listed in Fig. 1 for incident light linearly polarized along the [001] direction;  $\beta = \frac{2\hbar\omega}{(2n_R c \epsilon_0)^2} \xi^{xxxx}$ . Here  $n_R$  is the refractive index,  $c$  is the speed of light, and  $\epsilon_0$  is the vacuum permittivity. We restrict our calculations to photon energies below the onset of one-photon indirect absorption; beyond this energy divergences in Eq. (3) become problematic and a more sophisticated theory taking into account real population in intermediate states is necessary. Our results are consistent with the measured values of Bristow *et al.*<sup>15</sup> The calculated  $\beta$  at  $1.55 \mu\text{m}$  is around  $1.0 \text{ cm/GW}$ .

When the laser is incident along a direction other than [001], or is not linearly polarized, other  $\xi^{abcd}$  contribute to the absorption and lead to its anisotropy.<sup>16</sup> Fig. 2 (a) shows the spectra of  $\xi^{xxxx}$ ,  $\xi^{xxyy}$ , and  $\xi^{xyxy}$  at temperature 4 K (red thin curves) and 300 K (black thick curves).

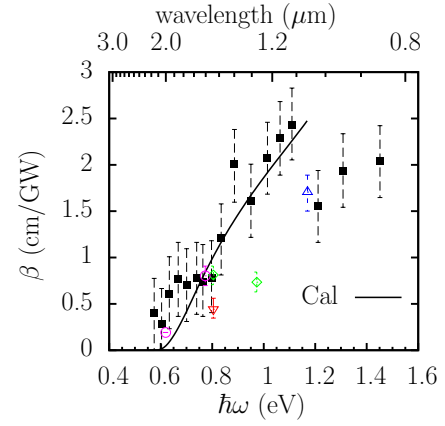


FIG. 1. (color online). Spectrum of  $\beta$  at 300 K. The circles,<sup>10</sup> uptriangles,<sup>12</sup> downtriangles,<sup>13</sup> diamonds,<sup>14</sup> and squares<sup>15</sup> are experimental results. The solid curve is our calculation.

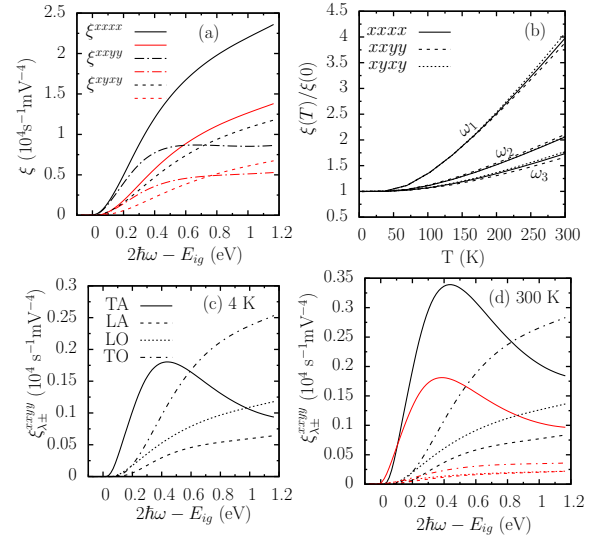


FIG. 2. (color online). Detailed calculation results. (a) Spectra of  $\xi^{xxxx}$ ,  $\xi^{xxyy}$ , and  $\xi^{xyxy}$  at 4 K (red thin curves) and 300 K (black thick curves). (b) Temperature dependence of  $\xi(T)/\xi(0)$  at different photon energies  $\hbar\omega_i$ :  $2\hbar\omega_i - E_{ig} = 0.1, 0.43, \text{ and } 1 \text{ eV}$  for  $\omega_1, \omega_2$  and  $\omega_3$  respectively; at 0 K  $\omega_2$  corresponds to a wavelength of  $1.55 \mu\text{m}$ . (c)-(d) Phonon-resolved spectra of  $\xi^{xxyy}$  at 4 K (c) and 300 K (d); black thick (red thin) curves are for phonon emission (absorption) processes.

When the excess photon energy is less than 1.2 eV, all injection coefficients increase with both photon energy and temperature, except that  $\xi^{xxyy}$  increases to a maximum value and then decreases slightly with photon energy at 300 K. Compared to the one-photon indirect injection coefficient, which is nearly quadratic in photon energy,<sup>23</sup> the two-photon indirect injection coefficients increase more slowly due to the explicit dependence on  $\omega^{-4}$  and the complicated frequency dependence of the transition matrix elements appearing in Eq. (3).

To better understand the spectra, in Fig. 2 we also

plot the phonon-resolved spectra of  $\xi_{\lambda+}^{xyxy}$  at 4 K (c) and 300 K (d) as black thick curves. The LA phonon assisted process is least important at all temperatures, and gives a contribution less than 15%; the TA and TO phonon assisted processes give the most important contributions, and dominate at low and high photon energies respectively. The crossover photon energy, where the TO phonon assisted process first dominates, moves to higher photon energy with increasing temperature because the TA phonon has the smaller energy, and the TA phonon assisted process is more sensitive to temperature. At 300 K, the spectra of  $\xi_{\lambda-}^{xxxx}$  are given as red thin curves in Fig. 2 (d). Only the TA phonon absorption assisted process gives a significant contribution. Therefore, the photon energy dependence of each absorption process is strongly determined by the properties of the mediated phonons, and selection rules taking into account the phonon properties are necessary.

In Fig. 2 (b) we give the temperature dependence of the injection coefficient at different photon energies. When the temperature is higher than 200 K, the injection coefficients depend approximately linearly on temperature, with the slope decreasing with photon energy. As with one-photon indirect injection,<sup>23</sup> the only temperature dependence comes from the equilibrium phonon number  $N_{q\lambda}$ , which is approximately linear with temperature at high temperatures.

In contrast to the two-photon direct absorption coefficient, which has a cusp<sup>30,31</sup> that identifies the onset of absorption from the spin-split off band, the spectrum of  $\beta$  in indirect absorption is very smooth. This is because the energy dependence at the onset of indirect absorption is  $\propto (\hbar\omega - E_{ig})^2$ , instead of  $\propto (\hbar\omega - E_g)^{1/2}$  for direct absorption. In contrast to one-photon indirect gap absorption,<sup>23</sup> the TA phonon assisted process here is at least as important as TO phonon assisted process.

In conclusion, we have performed a full band calculation of two-photon indirect absorption in bulk silicon. We find the two-photon indirect absorption coefficient is about 1 cm/GW at 1.55  $\mu\text{m}$  and 300 K. The TA and TO assisted processes give the most important contributions at all temperatures. The injection coefficients depend linearly on temperature over 200 K.

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